Response Surface Methodology Based on Central Composite Design for Investigation of the Mean Drop Behaviors in Reactive Extraction System with Presence of Uranium in a Horizontal Pulsed Column

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ABSTRACT: To carry out this investigation, drop behavior in a horizontal pulsed sieve-plated column was measured under with and without reactive extraction situations. Central composite design, a subcategory of response surface methodology, was utilized to survey the influence of the operational parameters on the drop behavior. The effect of the reactive extraction situation was also observed on droplet behavior. According to the experimental results, the effect of pulse intensity on the droplet behavior was greater than the phase flow rates. For the prediction of the Sauter mean drop diameter in a reactive extraction situation, a new correlation was determined. The results obtained via the proposed correlation were closely matched with the experimental results (AARE is about 6.64%). Also, to determine a predictive correlation for drop size distribution, log-normal, and normal probability density function were examined. The constant parameters at these probability density functions are specified by the obtained results as well as the mathematical approach.

KEYWORDS: Sauter Mean Drop Diameter; Probability Density Function; Horizontal Pulsed Column; Extraction Efficiency; Central Composite Design.

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INTRODUCTION

Uranium is a fundamental element in the nuclear fuel cycle, but its ores are very limited. This element is a radioactive material on ground and surface water that is very reactive. Therefore, the uranium extraction from the sources is essential [1]. The solvent extraction process generally is used for recovery of the hazardous metal from the waste. Hence, solvent extraction is a major technique for uranium extraction from the ores on the industrial scale [2-5]. Solvent extraction has various advantages, including effortless to handle, low cost at setup process, minimum waste production, product with high purity and maximum production capacity [6]. According the presented literature, it can be said that the tertiary amine extractants such as Alamine 336 are suitable for uranium extraction from sulfate solutions [7-10].

The solvent extraction method is an industrial method for the separation of elements. Thus, lately a large number of extractors widely have been investigated and developed [11]. In the liquid-liquid extractor system, the mass transfer rate is directly proportional to the interfacial area and degree of turbulence [12, 13]. It can be said that in terms of safety and cost-effectiveness, the pulse columns are preferable to other extractors. The main advantages of pulsed columns are: they have no moving parts, therefore, the cost of design, repair and maintenance in such columns is low. Furthermore, they are usable in corrosive materials and radioactive solutions. These advantages caused to the development of the pulsed columns for application in chemical, petrochemical and specially nuclear industries [14]. The pulsed columns are divided two categories: vertical and horizontal types. The horizontal and vertical columns are appropriate for industries that have height and surface area restrictions, respectively [14-18]. Although the vertical type has enough requirements for applications in the nuclear industry, the horizontal type can be replaced the vertical type according to additional advantages. The most important advantages of this type of column are: the decrease in building height, the reduction in the operating costs for shielding, the modification in the process control, availability of quick and easy column in the radioactive processes and the decrement in the risk of radioactive contamination of equipment [18].

The design of the extraction columns is very important to achieve the appropriate flow rates of two phases and mass transfer performance [2]. Knowledge of drop behavior is an essential parameter for the scale up and design of the extraction columns. It affects the residence time of the dispersed drops, the dispersed phase holdup and throughput of the column [19-21]. In the liquid-liquid extraction columns, an average volume surface diameter of the organic droplets is generally utilized to determine the contact times, interfacial area and mass transfer coefficient of both the continuous and dispersed phase [22, 23]. Therefore, the various drop size distributions which have different interfacial areas can be determined the same mean drop size in the extraction columns [20, 21, 24]. The changes of drop size distribution in the extraction column lead to a hold-up profile, which can be used to determine the column flooding [25]. The knowledge of drop behavior along the equipment as a function of the operational parameters and physical characteristics of the systems is fundamental to get a realistic explanation of column performance and to develop this column in the extraction process [26]. Many studies were carried out on drop behavior in the vertical columns [20, 27-30]. Nonetheless, for the horizontal columns, there are few reports about the droplet behavior in without reactive extraction situation [31, 32].

According to previous studies, there is few data for design and scale up of the horizontal pulsed columns under reactive extraction conditions. Also, due to the importance of droplets behavior in the extraction columns design, the aim of this study is measuring the droplet behavior and probability density function in this column with changing operating parameters (pulse intensity, flow rate of continuous and disperse phases). In this investigation, two different chemical systems were used to study the effect of the reactive extraction and without reactive extraction conditions on the droplet behavior. Furthermore, using the obtained experimental results, the empirical correlations for drop behavior were derived in the reactive extraction situation. It can be deduced that this column is a suitable option for the uranium extraction from a sulfate solution with about 99% efficiency.

EXPERIMENTAL SECTION

Equipment of pilot column

The horizontal pulsed sieve column was made of Pyrex glass, with a bore of 6.2 cm and with a length of 150 cm. The general specific geometries and schematic diagram of the mentioned column are shown in Fig. 1.

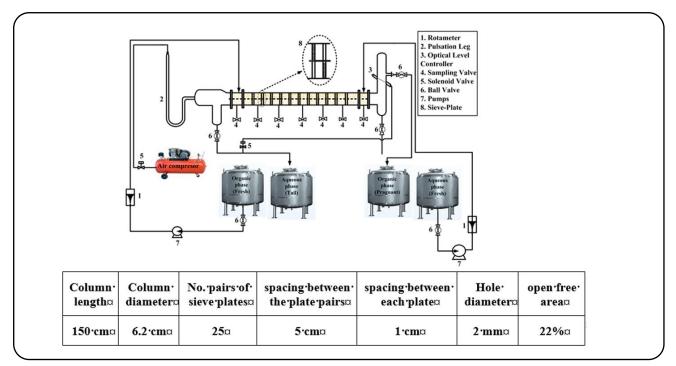


Fig. 1: Schematic diagram of the pilot plant horizontal pulsed sieve-plate extractor.

The active section of the column was containing 25 pairs of sieve plates that built of 304 stainless steel. The holes on the sieve plate near the inlets of dispersed and continuous phase were placed down and up, respectively. After the contact of two phases in the active section of the column, the dispersed and continuous phases are separated and collected in two settlers with 5 cm radius. Also, the aqueous and organic phase flow rates were measured by Rotameter. An air pulsing system created a pulse in the column. The interface of the two phases was controlled by an optical sensor that located at the top of the aqueous phase inlet and under the organic phase outlet in the upper settler.

Reagents

Commercial grade of Alamine 336 from NetSun Company of China and kerosene were used as an organic phase. Also, AR grade isodecanol from Merck was employed as a modifier. The leach liquor containing 250 mg/L uranium used in the reactive experimental system as an aqueous phase was generated in the Ghachin site of Bandar Abbas. Sulfuric acid with analytical grade as an aqueous phase was used in without reactive extraction situation. In this research work, two liquid-liquid system were utilized for experiments. The physical characteristics

of the two systems (with and without reactive extraction systems) are summarized in Table 1.

Analysis apparatus

Through the process of our experimentation, an Inductively Coupled Plasma Atomic Emission Spectrophotometer (ICP-AES Perkin-Elmer-7300 DV) was employed to determine the concentration of uranium in the continuous phase. The quantity of viscosity and interfacial tension were measured by DVI-Prime viscometer and a Krüss tensiometer, respectively. Also, the measurement of the solutions density data was carried out by the pycnometer method.

Procedure

The startup of the column in all the experimental runs without and with reactive extraction was far from flooding conditions. After filling the column with the continuous phase, the organic phase was entered to the column as dropwise. All experiments were done at the temperature up to $20\,^{\circ}\text{C}$.

In this study, Response Surface Methodology (RSM) with three input variables such as pulse intensity (at range 0.8-1.2 cm/s), continuous and dispersed phase flow rates (within the range 1-5 l/h) are used to study the effects

Table 1: Physical characteristics of the two systems used in this research.

properties	Phase	Viscosity (×10 ³ kg/m.s)	Density (kg/m³)	Interfacial tension (×10³ N/m)
With reactive extraction	Sulfate leach liquor (as continuous phase)	0.86	1058	9.9
	Saturated organic phase (as dispersed phase)	1.84	820	9.9
Without reactive extraction	Diluted sulfuric acid (as continuous phase)	0.76	1009	12.2
without reactive extraction	Alamine, isodecanol and kerosene (as dispersed phase)	1.52	803	12.2

Table 2: The CCD matrix and the response values for two systems.

Run $Q_c(L/h)$ $Q_d(L/h)$	O. (I /b)	af (cm/sec)	Without reactive extraction system	With reactive extraction system		
	ai (ciii/sec)	d ₃₂ (mm) (Exp.)	d ₃₂ (mm) (Exp.)	E _U (%) (Exp.)		
1	4	2	0.9	1.29	1.14	94
2	3	3	1	1.13	0.98	97.5
3	4	4	0.9	1.42	1.31	95.4
4	4	2	1.1	0.87	0.73	97.8
5	3	3	1.2	0.64	0.45	99.16
6	3	3	1	1.14	0.95	97.35
7	2	2	1.1	0.77	0.62	98.1
8	3	3	1	1.12	0.99	97.6
9	1	3	1	1.06	0.92	98.46
10	2	4	1.1	0.87	0.69	98.96
11	3	5	1	1.30	1.11	98.75
12	3	3	1	1.11	0.97	97.65
13	2	2	0.9	1.19	1.04	95.1
14	5	3	1	1.24	1.19	96.5
15	3	1	1	1.07	0.92	96.72
16	3	3	0.8	1.55	1.44	93.42
17	3	3	1	1.15	0.98	97.2
18	4	4	1.1	0.97	0.82	98.3
19	2	4	0.9	1.36	1.18	97.4
20	3	3	1	1.14	0.99	97.8

on Sauter mean drops diameter (d_{32}) , drops distribution (P_n) and uranium extraction efficiency (%E_u). Design Expert software 7.0 was used to design the experiments. The statistical testing is done by ANOVA analysis with p-Value, R-Squared, adjusted R-Squared and predicted R-Squared. The operating conditions as suggested by Central Composite Design and responses at each run are reported in Table 2.

Measurement of drop diameter

After reaching steady-state situations at the end of each experimental run, the photographic method with a Nikon digital camera (D5000 model) was applied for measurement of the drop diameters. The drop diameter was measured by analyzing the recorded photos with AutoCAD 2016 software. The Sauter mean drop diameter (d₃₂) is determined as follows:

$$d_{32} = \frac{\sum_{i=0}^{N} n_i d_i^3}{\sum_{i=0}^{N} n_i d_i^2}$$
 (1)

where n_i is the numbers of droplets and d_i is mean diameter within range i.

Measurement of extraction efficiency

At the steady-state conditions with Sampling from the seven valves installed in 25 cm distances on the column and determination of uranium in samples, the uranium extraction efficiency at the end of each run is calculated by the following equation:

Uranium Extraction Efficiency (%) =
$$\frac{x_1 - x_7}{x_1 - x_7} \times 100$$
 (3)

Where x_1 and x_7 are uranium concentrations of the aqueous phase at input and output valve streams. Also, x_7^* is the equilibrium uranium concentration of the aqueous phase across the output valve stream. The uranium concentration in the aqueous phase at the column length (x_1 to x_7) was measured by ICP-AES spectrophotometer.

RESAULTS AND DISCUSSION

Variation of sauter mean drop diameter

Effects of Operating Parameters

The results of ANOVA for the Sauter mean drop diameter in two systems are presented in Table 3. The p-value is <0.001 that obviously confirms the experimental results. The value of correlation coefficient "R-square", "adjusted R-square" and "predicted R-square" shows the accuracy of the experiment design within two systems.

The effect of changing operational parameters on the Sauter mean drop diameter is presented in Fig. 2. At all flow rates of the continuous phase as shown in Fig. 2(a), the Sauter mean drop diameter significantly decreases with increasing pulse intensity. An increase in the pulse intensity gave rise to the interfacial forces of the dispersed phase drops, therefore, the breakage of drops occurred and consequently the Sauter mean drop diameter decreased along the column. At all the pulse intensity, a smooth increase in the Sauter mean diameter of the drop is observed with the increase in flow rates of the continuous phase. As shown in Fig. 2(b) at all the pulse intensity, the Sauter mean drop diameter slightly increases with the increase in the dispersed phase flow rates. The effect of the dispersed and continuous phase flow rates on Sauter mean drop diameter are shown in Fig. 2(c) for the two systems.

It should be noted that a slight increase in the Sauter mean drop diameter is observed with the increase in flow rates of the dispersed and continuous phase. Therefore, it is observed from these figures that the effect of pulsing intensity is stronger than that of the phase flow rate on the Sauter mean drop diameter. As shown in Fig. 2, the Sauter mean drop diameter in without reactive extraction system is larger than that the with reactive extraction system because the interfacial tension in without reactive extraction system is higher than the other system.

New correlation for prediction of sauter mean diameter of drop

Many correlations have been suggested for determination of the Sauter mean diameter of drop in various vertical pulsed extractors [27]. Nevertheless, these correlations do not have enough accuracy to predict Sauter mean diameter of drops and cannot be used in the horizontal pilot plant columns and the reactive extraction situation.

The comparison of the experimental results of this investigation with the values obtained from the empirical correlation recommended in previous works. The AARE values between experimental results of this investigation and the suggested equations by *Khajenoori et al.* and *Panahinia et al.* were 38.51 and 30.80%, respectively. Therefore, the previous correlations are not proper to predict the mean drop size because the mentioned equation doesn't consider the reaction situations, whereas the reaction between uranium and Alamine 336 is included in the experimental data. Consequently, a novel modified correlation with consideration of the reactive extraction situation is derived by using the Buckingham's pi-theorem as follows:

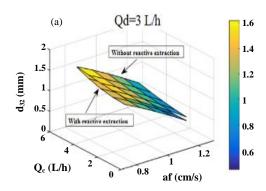
$$\frac{d_{32}}{\left(\frac{Q_c \cdot \rho_c}{\mu_c}\right)} = 3.07 \times 10^4 \left(\frac{Q_c \cdot \rho_c^2 \cdot af}{\mu_c^2}\right)^{-1.95}$$

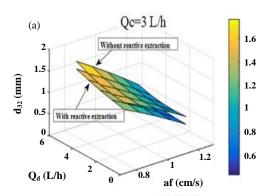
$$\left(2 + \frac{Q_d}{Q_c}\right)^{0.51} \left(\frac{\rho_d}{\rho_c}\right)^{1.67.62} \left(\frac{\mu_d}{\mu_c}\right)^{25.61} \left(\frac{Q_c \cdot \rho_c^2 \cdot \sigma}{\mu_c^3}\right)^{1.27}$$

The AARE between the produced results by Eq. (5) and experimental results for this research column is 6.64%. Therefore, the experimental and predicted data were near enough to the bisector line which confirms the accuracy of the proposed correlation.

Table 3: The ANOVA to RSM variables for the Sauter mean drop diameter in two systems.

Source	Sum of Squares	Degree freedom	F Value	p-Value
	Without r	eactive extraction system		
Model	0.91	9	71.16	< 0.0001
A-af	0.81	1	570.03	< 0.0001
B-Qc	0.032	1	22.31	0.0032
C-Qd	0.058	1	40.77	0.0007
AB	1.96×10 ⁻⁴	1	0.14	0.7229
AC	1.447×10 ⁻³	1	1.02	0.3521
BC	2.86×10 ⁻⁴	1	0.20	0.6696
A^2	6.556×10 ⁻⁴	1	0.46	0.5226
B^2	6.578×10 ⁻⁴	1	0.46	0.5219
C^2	3.885×10 ⁻³	1	2.73	0.1495
Residual	8.536×10 ⁻³	6	-	-
Lack of Fit	8.374×10 ⁻³	5	10.31	0.2319
Pure Error	1.624×10 ⁻³	1	-	-
Cor Total	0.92	15	-	-
R-Squared	0.9907	-	-	-
Adj R-Squared	0.9768	-	-	-
Pred R-Squared	0.9283	-	-	-
	With rea	active extraction system	1	
Model	0.96	9	37.49	0.0001
A-af	0.84	1	295.67	< 0.0001
B-Qc	0.055	1	19.50	0.0045
C-Qd	0.052	1	18.36	0.0052
AB	3.381×10 ⁻⁴	1	0.12	0.7416
AC	1.314×10 ⁻³	1	0.46	0.5213
ВС	6.301×10 ⁻⁶	1	2.223×10 ⁻³	0.9639
A^2	5.484×10 ⁻⁴	1	0.19	0.6754
B^2	4.703×10 ⁻³	1	1.61	0.2451
C^2	7.72×10 ⁻⁴	1	0.27	0.6204
Residual	0.017	6	-	-
Lack of Fit	0.017	5	70.94	0.0899
Pure Error	4.78×10 ⁻⁵	1	-	-
Cor Total	0.97	15	-	-
R-Squared	0.9825	-	-	-
Adj R-Squared 0.9563		-	-	-
Pred R-Squared	0.8615	-	_	-





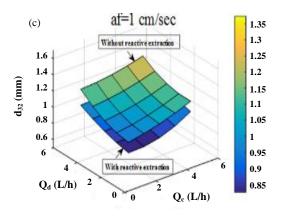


Fig. 1: The effect of operational parameters on the Sauter mean drop size for two systems.

Variation of drop size distribution

Effects of operating parameters

In this section, the effect of the operational parameters such as the pulse intensity and flow rate of the phases on the drop size distribution was investigated. The various pulse intensities on the drop size distribution at the constant flow rates of continuous and dispersed phases for the two systems were surveyed that results are presented in Fig. 3.

With more turbulent conditions, homogeneous distributions and smaller droplets are generated for two systems. Thus, the increasing pulse intensity leads to a narrower and more symmetric distribution around the mean drop size value. Also, the distributions of drop size shift to the left within both systems. The influence of various dispersed and continuous phase flow rates on the drop size distribution for the two systems was investigated, the results of which are shown in Fig. 4. As can be observed in this figure, with increasing in both phase flow rates, the distribution shape of drop size in these systems shifts to the right. But, with changing these parameters without mass transfer system, the drop size distribution shifts towards larger drops. This phenomenon can be explained by the existence of the higher interfacial tension and the absence of reactive extraction in this system.

New correlation for prediction of drop size distribution

As shown in the previous studies [23, 33-35], the probability density of drops used for predicting of drop size distribution in liquid–liquid extractors. The ratio of the droplets number with a specific diameter to the total droplets number is called the probability density of droplets. The analysis of the drop size distribution is often done by log-normal and normal probability density functions. Therefore, the mentioned probability density functions (Eqs. (6) and (7)) were investigated to predict the experimental data on the drop size distributions.

$$P_{n}(d) = \frac{1}{\sqrt{2\pi\alpha}} exp \left[-\left(\frac{(d-\beta)}{\sqrt{2\alpha}}\right)^{2} \right]$$
 (6)

$$P_{1g}(d) = \frac{1}{\sqrt{2\pi d \alpha}} exp \left[\left(\frac{(\ln d - \beta)}{\sqrt{2\alpha}} \right)^{2} \right]$$
 (7)

Where d_i is the drop diameter and α and β denote the constant parameters. In order to predict the drop size distribution, a nonlinear regression analysis was used to fit these functions to the experimental results, then the constant parameters for these functions are determined.

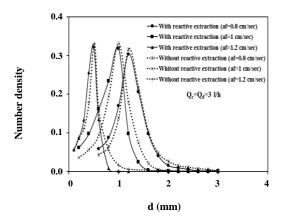
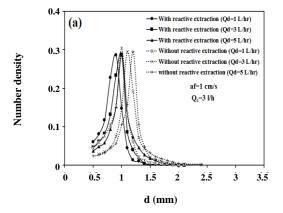


Fig. 3: The effect of pulse intensity on the drop size distribution.



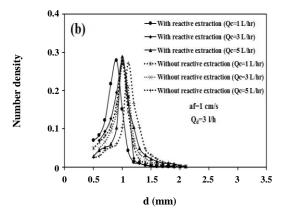


Fig. 4: The effect of dispersed and continuous phase flow rates on the drop size distribution. (a): various dispersed phase flow rate. (b): various continuous phase flow rate.

In addition, the new correlations for the evaluated parameters with consideration of the reactive extraction conditions are derived by using the Buckingham's pi-theorem based on physical characteristics and operational parameters as follows:

$$\alpha = 7.89 \times 10^{-7} \left(\frac{Q_c \cdot \rho_c^2 \cdot af}{\mu_c^2} \right)^{1.054} \left(2 + \frac{Q_d}{Q_c} \right)^{-0.206} \left(\frac{\rho_d}{\rho_c} \right)^{-35.58} \left(\frac{\mu_d}{\mu_c} \right)^{19.9} \left(\frac{Q_c \cdot \rho_c^2 \cdot \sigma}{\mu_c^3} \right)^{-1.15}$$
(8)
$$\beta = 6.75 \times 10^{-8} \left(\frac{Q_c \cdot \rho_c^2 \cdot af}{\mu_c^2} \right)^{1.206} \left(2 + \frac{Q_d}{Q_c} \right)^{0.035} \left(\frac{\rho_d}{\rho_c} \right)^{-35.56} \left(\frac{\mu_d}{\mu_c} \right)^{19.87} \left(\frac{Q_c \cdot \rho_c^2 \cdot \sigma}{\mu_c^3} \right)^{-1.103}$$
(9)
$$\alpha = 3.98 \times 10^4 \left(\frac{Q_c \cdot \rho_c^2 \cdot af}{\mu_c^2} \right)^{2.03} \left(2 + \frac{Q_d}{Q_c} \right)^{-0.132} \left(\frac{\rho_d}{\rho_c} \right)^{348.84} \left(\frac{\mu_d}{\mu_c} \right)^{77.47} \left(\frac{Q_c \cdot \rho_c^2 \cdot \sigma}{\mu_c^3} \right)^{1.96}$$
(10)

$$\beta = 3.96 \times 10^4 \left(\frac{Q_c \cdot \rho_c^2 \cdot af}{\mu_c^2}\right)^{1.55} (2 + \frac{Q_d}{Q_c})^{0.41} \left(\frac{\rho_d}{\rho_c}\right)^{265.58} \left(\frac{\mu_d}{\mu_c}\right)^{48.92} \left(\frac{Q_c \cdot \rho_c^2 \cdot \sigma}{\mu_c^3}\right)^{1.81}$$
(11)

Eqs. (8) and (9) and Eqs. (10) and (11) depict the constant parameters of the log-normal and normal functions, respectively. The AARE values to estimation of α and β parameters for log-normal and normal distribution functions are equal to 6.54, 9.28, 6.78 and 10.35 %, respectively. The results of comparison between these correlations and the present experimental investigation outcomes are depicted in Fig. 5. It is observed that these probability density functions are closely matched with the experimental results.

Extraction of uranium

In Eq. (3), the equilibrium uranium concentration is necessary for the determination of uranium extraction efficiency. As shown in Fig. 6, the uranium extraction isotherm with different ratios of the aqueous and organic phases (such as A/O= 40, 20, 10, 5, 2, 1 and 0.5) and constant concentrations of uranium in the aqueous phase (250 mg/L) was drawn. Also, the extraction efficiency against the dimensionless length of the column are depicted in Fig. 7.

Effects of operating parameters on the extraction efficiency

The results of ANOVA with the reactive extraction system are shown in Table 4. The p-value is <0.001 that obviously confirms the experimental results. The value of correlation coefficient "R-square", "adjusted R-square" and "predicted R-square" shows the accuracy of the experiment design within two systems.

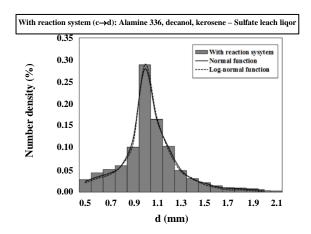


Fig. 5: Comparison of drop size distribution between experimental results and theoretical distribution functions for reaction system (af=1cm/s, Q_c =5 l/h, Q_d =3 l/h).

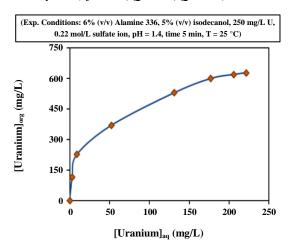


Fig. 6: Uranium extraction isotherm from sulfate solution using Alamine 336.

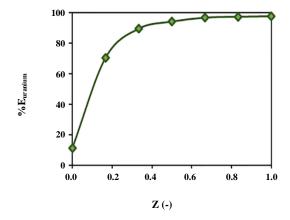


Fig. 7: Uranium extraction efficiency against dimensionless column length (af=1 cm/s, $Q_c=Q_d=3$ L/h).

In Fig. 8, the effect of altering operational conditions on the extraction efficiency is presented. The influence of the pulse intensities and continuous phase flow rates on the uranium extraction efficiency is illustrated in Fig. 8(a). The extraction efficiency significantly increases with increasing pulse intensity, at all continuous flow rates. The increasing of the pulse intensity is led to the variation of the drop sizes and holdup with axial mixing. Therefore, the mass transfer performance increases at this column. As shown in Fig. 8(a), with an increase in the energy supplied via pulse intensity and the breakage of the drops, the drop size decreases. The formation of smaller drops leads to turbulence in the droplet and increases the interfacial area between the two phases. Therefore, the extraction efficiency of uranium increases. With increasing continuous phase flow at all pulse intensity, the mean drop size increases in the extraction column. Therefore, the residence time of droplets is reduced and the droplets passed the column length rapidly. Consequently, the uranium extraction efficiency reduces with increasing the continuous phase flow rate. The influence of the dispersed phase flow rate and pulse intensity on the uranium extraction efficiency is presented in Fig. 8(b). As depicted in this figure, by increasing the dispersed phase flow rate, the fresher organic phase contacts the leach liquor solution. Therefore, at all pulse intensity, an increase the dispersed phase flow rate caused to the increase in the uranium extraction efficiency. The influence of the phase flow rates on the variation of uranium extraction efficiency is illustrated in Fig. 8(c). At all flow rates of the continuous phase as shown this figure, the increasing the flow rates of the dispersed phase leads to the increase the uranium extraction efficiency. Though, the uranium extraction efficiency decreases with increasing the flow rates of the continuous phase at all flow rates of the dispersed phase.

CONCLUSIONS

In the present work, variations of operational parameters by using the central composite design method on the drop behavior and extraction efficiency of uranium in a horizontal pulsed sieve-plate extractor were comprehensively studied. The results achieved from the experimental data were drawn:

-The relation between the Sauter mean drop diameter and uranium extraction efficiency are directly proportional to the pulse intensity.

Source	Sum of Squares	df	Mean Square	F Value	p-Value
Model	45.48	9	5.05	44.52	< 0.0001
A-af	32.21	1	32.21	283.75	< 0.0001
B-Qc	3.98	1	3.98	35.07	0.0010
C-Qd	5.20	1	5.20	45.80	0.0005
AB	0.57	1	0.57	5.04	0.0658
AC	0.68	1	0.68	6.03	0.0494
ВС	0.20	1	0.20	1.75	0.2342
A^2	1.61	1	1.61	14.21	0.0093
B^2	4.9×10 ⁻³	1	4.9×10 ⁻³	0.043	0.8423
C^2	0.034	1	0.034	0.30	0.6027
Residual	0.68	6	0.11		
Lack of Fit	0.68	5	0.14	27.04	0.1449
Pure Error	0.005	1	0.005		
Cor Total	46.16	15			
R-Squared	0.985246				
Adj R-Squared	0.963115				
Pred R-Squared	0.884835				

Table 4: The ANOVA to RSM variables for uranium efficiency in reactive extraction system.

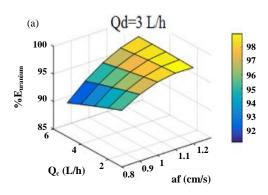
- A uniform drop size distribution and smaller drop size achieved because of the increasing the pulse intensity.
- The dispersed and continuous phase flow rates slightly affect the drop behavior.
- With increasing continuous phase flow rate, uranium extraction efficiency from the sulfate leach liquor by using Alamine 336 initially reduced, but increased with increasing dispersed phase flow rate.
- In the reactive extraction system, the interfacial tension influenced on drop size. In addition, the lognormal and normal distribution functions have a good agreement with these experimental results to predict the drop size distribution.
- The present work has provided useful information on the uranium extraction efficiency (approximately %99) and the drop behavior in a pilot plant horizontal pulsed sieve-plate column. The mentioned column can be applied in the extraction process in the nuclear industries. Moreover, in places with limited height, this column is a suitable alternative in radioactive processes.

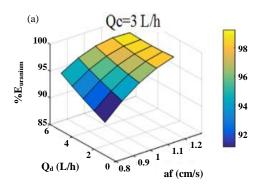
Nomenclature

AARE	Average absolute relative error
CCD	Central composite design
RSM	Response surface methodology
af	Pulse intensity (m/s)
Q	Volumetric flow rate (m ³ /s)
d_{32}	Sauter mean drop diameter (m)
n_i	Number of droplets of mean diameter di
P	Probability of number density

Greek letters

X	Mass fraction of uranium in the continuous phase
y	Mass fraction of uranium in the dispersed phase
X	Dimensionless solute concentrations of
	the continuous phase
Y	Dimensionless solute concentrations of
	the dispersed phase
Z	Column length (cm)
Z	Dimensionless length of the column
ρ	Density (kg/m³)
μ	Viscosity (Pa s)





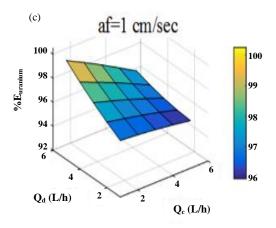


Fig. 8: The effect of operational parameters on the uranium extraction efficiency in with reaction system.

 σ Interfacial tension (N/m) α Constant parameter of probability of density function Constant parameter of probability of density function

Subscript aq Aqueous phase org Organic phase c Continuous phase d Dispersed phase * Equilibrium value n Normal probability density function

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Log-normal probability density function

REFERENCES

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- [1] Kumar J. R., Kim J., Lee J., Yoon H., A Brief Review on Solvent Extraction of Uranium From Acidic Solutions, Sep. & Purify. Reviews, 40: 77-125 (2011).
- [2] Rydberg J., Musikas C., Choppin G. R., "Solvent Extraction Principles Practice", CRC Press, New York, (2004).
- [3] Kislik V. S., Advances in Development of Solvents for Liquid-Liquid Extraction, "Solvent Extraction Classical and Novel Approaches", Elsevier (2012).
- [4] Khanramaki F., Shirani A.S., Safdari J., Torkaman R., Investigation of Liquid Extraction and Thermodynamic Studies on Uranium from Sulfate Solution by Alamine 336 as an Extractant, *International Journal of Environmental Science And Technology*, 15: 1467-1476 (2017).
- [5] Khanramaki F., Safdari J., Shirani A. S., Torkaman R., Investigations on the Complete Removal of Iron(Iii) Interference on the Uranium(Vi) Extraction from Sulfate Leach Liquor Using Alamine 336 in Kerosene, Radiochimica Acta, 106: 631-643 (2018).
- [6] Benedict M., Pigford T. H., H. W. Levi, Nuclear Chemical Engineering, 2nd ed., Mc Graw Hill Book Company, (1981).
- [7] Behera P., Mishra R., Chakravortty V., Solvent Extraction of Uranium (Vi) and Molybdenum (Vi) by Alamine 310 and Its Mixtures from Aqueous H₃PO₄ Solution, *J. Radioanal. Nucl. Chem.*, 173: 161-169 (1993).
- [8] Ramadevi G., Sreenivas T., Navale A.S., Padmanabhan N.P.H., Solvent Extraction of Uranium from Lean Grade Acidic Sulfate Leach Liquor with Alamine 336 Reagent, J. Radioanal Nucl. Chem., 294: 13-18 (2012).

- Khanramaki F. et al.
- [9] Quinn J.E., Wilkins D., Soldenhoff K.H., Solvent Extraction of Uranium from Saline Leach Liquors Using Dehpa/Alamine 336 Mixed Reagent, Hydrometallurgy, 134: 74-79 (2013).
- [10] Zahakifar F., Charkhi A., Torab-Mostaedi M., Davarkhah R., Kinetic Study of Uranium Transport via a Bulk Liquid Membrane Containing Alamine 336 as a Carrier, Journal of Radioanalytical and Nuclear Chemistry, 316: 247-255 (2018).
- [11] Godfrey J.C., Slater M.J., "Liquid-Liquid Extraction Equipment", New York: John Wiley & Sons, Inc. (1994).
- [12] Jaradat M., Attarakih M., Bart H., Population Balance Modeling of Pulsed (Packed and Sieve-Plate) Extraction Columns: Coupled Hydrodynamic and Mass Transfer, Ind. Eng. Chem. Res., 50: 14121-14135 (2011).
- [13] Samani G., Safdari J., Haghighi-Asl A., Torab-Mostaedi M., Effect of Structural Parameters on Drop Size Distribution in Pulsed Packed Columns, Chem. Eng. Technol., 37: 1155-(2014).
- [14] Hussain A.A., Liang T.-B., Slater M.J., Characteristic Velocity of Drops in a Liquid-Liquid Extraction Pulsed Sieve Plate Column, Chem. Eng. Res. Des., **66**: 541-554 (1988).
- [15] Brauer H., Sucker D., Biological Waste Water Treatment in a High-Efficiency Reactor, Ger. Chem. Eng., 2: 77-(1979).
- [16] Procházka J., Hafez M.M., The Analysis of the Dynamic Effects in Vibrating and Pulse Plate Extraction Columns, Collect. Czech. Commun., 37: 3725-3734 (1972).
- [17] Logsdail D.H., Thornton J.D., Developments in Horizontal Pulsed Contactors for Liquid-Liquid Extraction Processes, Journal of Nuclear Energy, 1: 15-(1959).
- [18] Melnyk A.J., Hydrodynamic Behavior of a Horizontal Pulsed Solvent Extraction Column, Part 1: flow Characterization, Throughput Capacity and Holdup, Can. J. Chem. Eng. J., 70: 417-(1992).
- [19] Maaß S., Wollny S., Voigt A., Kraume M., Experimental Comparison of Measurement Techniques for Drop Size Distributions Liquid/Liquid Dispersions, Exp. Fluids, 50: 259-269 (2011).

- [20] Asadollahzadeh M., Shahhosseini S., Torab-Mostaedi M., Ghaemi A., Drop Behavior in a Pilot Plant Oldshue-Rushton Extraction Column for Three Various Liquid-Liquid Systems, Separation and Purification Technology, 159: 7-17 (2016).
- [21] Thornton J.D., "Science and Practice Of Liquid-Liquid Extraction", University Press, Oxford (1992).
- [22] Moreira E., Pimenta L.M., Carneiro L.L., Faria R.C.L., Mansur M.B., Ribieron J.P., Hydrodynamic Behavior of Rotating Disc Contactor Under Low Agitation Conditions, Chem. Eng. Commun., 192(8): 1017-1035 (2007).
- [23] Oliveira N.S., Silva D.M., Gond M.P.C., Mansur M.B., A Study of The Drop Size Distributions and Hold-Up in Short Kühni Columns, Braz. J. Chem. Eng., 25(4): 729-(2008).
- [24] Ashrafmansouri S.S., Esfahany M.N., The Influence of Silica Nanoparticles on Hydrodynamics and Mass Transfer in Spray Liquid-Liquid Extraction Column, Sep. Purify. Technol., 151: 74-81 (2015).
- [25] Ousmane S., Isabelle M., Mario M., Mamadou T., Jacques A., Study of Mass Transfer and Determination of Drop Size Distribution in Extraction Column, Chem. Eng. Res. Des., 89: 60-68 (2011).
- [26] Grinbaum B., The Existing Models for Simulation of Pulsed and Reciprocating Columns - How Well Do They Work in The Real World?, Solv. Extr. and Ion Exch., 24: 795-822 (2006).
- [27] Yadav R.L., Patwardhan A.W., Design Aspects of Pulsed Sieve Plate Columns, Chem. Eng. J., 138: 389-415 (2008).
- [28] Usman M.R., Sattar H., Hussain S.N., Muhammad H., Asghar A., Afzal W., Drop Size in a Liquid Pulsed Sieve-Plate Extraction Column, Braz. J. Chem. Eng., **26**: 677-(2009).
- [29] Samani M.G., Haghighi-Asl A., Safdari J., Torab-Mostaedi M., Drop Size Distribution and Mean Drop Size in a Pulsed Packed Extraction Column, Chem. Eng. Res. Des., 90: 2148-2154 (2012).
- [30] Torab-Mostaedi M., Ghaemi A., Asadollahzadeh M., Flooding and Drop Size in a Pulsed Disc and Doughnut Extraction Column, Chemical Engineering Research and Design, 89: 2742-2751 (2011).

- [31] Khajenoori M., Haghighi-Asl A., Safdari J., Mallah M.H., Prediction of Drop Size Distribution in a Horizontal Pulsed Plate Extraction Column, *Chemical Engineering and Processing*, **92**: 25-32 (2015).
- [32] Panahinia F., Ghannadi-Maragheh M., Safdari J., Amani P., Mallaha M.H., Experimental Investigation Concerning the Effect of Mass Transfer Direction on Mean Drop Size and Holdup in a Horizontal Pulsed Plate Extraction Column, *Royal Society of Chemistry, RSC Advances*, 7: 8908-8921 (2017).
- [33] Rincon-Rubio L.M., Kumar A., Hartland S., Drop Size Distribution and Average Drop Size in a Wirz Extraction Column, *Chem. Eng. Res. Des.*, **72**: 493-502 (1994).
- [34] Zhang S.H., Yu S.C., Zhou Y.C., Su Y.F., A Model for Liquid–Liquid Extraction Column Performance the Influence of Drop Size Distribution on Extraction Efficiency, *Can. J. Chem. Eng.*, **63**: 212-226 (1985).
- [35] Chen W.L., Zhong L.M., Chen L., Weiyang W., Measurement and Analysis of Bimodal Drop Size Distribution in a Rotor–Stator Homogenizer, *Chem. Eng. Sci.*, **102**: 622-631 (2013).